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EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES. LIQUEFACTION: SECTION 3. H-COAL PROCESS

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Exxon Research and Engineering Company

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The report gives results of a review of the H-Coal Process of Hydrocarbon Research, Inc., from the standpoint of its effect on the environment. Quantities of solid, liquid, and gaseous effluents are specified, where possible, as well as the thermal efficiency of the process. Techniques for controlling pollution are outlined and discussed. For the purpose of reducing environmental impact, a number of possible modifications or alternatives are presented for consideration. In some areas existing information or control systems are inadequate; therefore, technology needs are pointed out covering such areas, together with approaches to improve efficiency and conservation of energy or water.

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LIQUEFACTION: SECTION 3. H-COAL PROCESS

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TABLE OF CONTENTS

		Page
1.	SUMMARY	1
2.	INTRODUCTION	2
3.	PROCESS DESCRIPTION	4
	3.1 Coal Preparation and Feeding	4
	3.2 Liquefaction Section	4
	3.3 Gas Separation and Cleanup	6
	3.4 Liquid Product Recovery	6
	3.5 Hydrogen Manufacture	7
	3.6 Auxiliary Facilities	7
4.	EMISSIONS TO ATMOSPHERE	12
	4.1 Coal Preparation and Feeding	12
	4.2 Liquefaction Section	24
	4.3 Gas Separation and Cleanup	24
	4.4 Liquid Product Recovery	25
	4.5 Hydrogen Manufacture	26
	4.6 Auxiliary Facilities	28
5.	EFFLUENTS - LIQUIDS AND SOLIDS	32
	5.1 Coal Preparation and Feeding	32
	5.2 Liquefaction Section	32
	5.3 Gas Separation and Cleanup	33
	5.4 Liquid Product Recovery	34
	5.5 Hydrogen Manufacture	35
	5.6 Auxiliary Facilities	36
6.	SULFUR BALANCE	39
7.	THERMAL EFFICIENCY	41
8.	TRACE ELEMENTS	43
9.	TECHNOLOGY NEEDS	47
10.	PROCESS DETAILS	51
11.	QUALIFICATIONS	56
12.	BIBLIOGRAPHY	57

LIST OF TABLES

No.		Page
1	Inputs to H-Coal Plant.	10
2	Outputs from H-Coal Plant	11
3	Description of Streams for H-Coal Liquefaction Plant.	14
4	Sulfur Balance H-Coal Plant	40
5	Thermal Efficiency H-Coal Plant	42
6	Example of Trace Elements That May Appear in Gas Cleaning Systems.	45
7	Technology Needs.	48
8	Steam Balance H-Coal Plant.	52
9	Electric Power Required in H-Coal Plant	53
10	Water Balance for H-Coal Plant.	53
11	Make Up Chemicals and Catalyst Requirements	54
12	Potential Odor Emissions.	55
13	Potential Noise Problems.	55

LIST OF FIGURES

No.		Page
1	Block Flow Plan for H-Coal Plant for Coal Liquefaction.	5
2	Effluents and Emissions from H-Coal Liquefaction Plant	13

TABLE OF CONVERSION UNITS

To Convert From	To	Multiply By
Btu	Calories, kg	0.25198
Btu/pound	Calories, kg/kilogram	0.55552
Cubic feet/day	Cubic meters/day	0.028317
Feet	Meters	0.30480
Gallons/minute	Cubic meters/minute	0.0037854
Inches	Centimeters	2.5400
Pounds	Kilograms	0.45359
Pounds/Btu	Kilograms/calorie,kg	1.8001
Pounds/hour	Kilograms/hour	0.45359
Pounds/square inch	Kilograms/square centimeter	0.070307
Tons	Metric tons	0.90719
Tons/day	Metric tons/day	0.90719

1. SUMMARY

The H-Coal Process of Hydrocarbon Research Inc., is reviewed from the standpoint of its effect on the environment. Quantities of solid, liquid and gaseous effluents are specified where this is possible, as well as the thermal efficiency of the process. Techniques for controlling pollution are outlined and discussed. For the purpose of reducing environmental impact, a number of possible modifications or alternatives are presented for consideration. In some areas existing information or control systems are inadequate, therefore technology needs are pointed out covering such areas, together with approaches to improve efficiency and conservation of energy or water.

2. INTRODUCTION

Along with improved control of air and water pollution, the country is faced with urgent needs for energy sources. To improve the energy situation, intensive efforts are under way to upgrade coal, the most plentiful domestic fuel, to solid, liquid and gaseous fuels which give less pollution. Other processes are intended to convert liquid fuels to gas. A few of the coal gasification processes are already commercially proven, and several others are being developed in large pilot plants. These programs are extensive and will cost millions of dollars, but this is warranted by the projected high cost for commercial gasification plants and the wide application expected in order to meet national needs. Coal conversion is faced with potential pollution problems that are common to coal-burning electric utility power plants in addition to pollution problems peculiar to the conversion process. It is thus important to examine alternative conversion processes from the standpoint of pollution and thermal efficiencies, and these should be compared with direct coal utilization when applicable. This type of examination is needed well before plans are initiated for commercial applications. Therefore, the Environmental Protection Agency arranged for such a study to be made by Exxon Research and Engineering Company under Contract No. EPA-68-02-0629, using all available nonproprietary information.

The present study under the contract involves preliminary design work to assure that the processes are free from pollution where pollution abatement techniques are available, to determine the overall efficiency of the processes, and to point out areas where present technology and information are not available to assure that the processes are nonpolluting. This is one of a series of reports on different fuel conversion processes.

All significant input streams to the processes must be defined, as well as all effluents and their compositions. This requires complete mass and energy balances to define all gas, liquid, and solid streams. With this information, facilities for control of pollution can be examined and modified as required to meet environmental objectives. Thermal efficiency is also calculated, since it indicates the amount of waste heat that must be rejected to ambient air and water and is related to the total pollution caused by the production of a given quantity of clean fuel.

Suggestions are included concerning technology gaps that exist for techniques to control pollution or conserve energy. Maximum use was made of the literature and information available from developers. Visits and/or contacts were made with the developers to update published information. Not included in this study are such areas as cost, economics, operability, etc. Coal mining and general offsite facilities are not within the scope of this study.

Previous studies in this program to examine environmental aspects of fossil-fuel conversion processes covered various methods for gasifying coal to make synthetic natural gas or low Btu gas. Reports have been issued on the Koppers, Synthane, Lurgi, CO_2 Acceptor, Bigas, Hygas, U-Gas and Winkler processes (1,2,3,4,5,6,7,8). In addition, an environmental study was made of the Meyers Process, which uses extraction to remove inorganic sulfur from coal (9).

In the area of coal liquefaction, reports have been issued on the COED Process of FMC (10) to make gas, tar, and char, as well as on the SRC Process of Pittsburg & Midway Coal Mining Company to make a heavy liquid clean boiler fuel (11).

These studies have now been extended to include coal liquefaction using the H-Coal process being developed by Hydrocarbon Research Inc. The present report covers our evaluation of environmental aspects of the H-Coal process. Considerable information is available in the literature on the products from the process as well as raw materials consumed, together with their properties and compositions (12,13,14,15,16). Our study is based primarily on reference 15, for the case making synthetic crude from Illinois coal, using 18,600 SCF of hydrogen per ton of coal. As in previous studies of this series, a complete and self-sufficient plant has been defined, avoiding for example purchased power which would cloud the basis for evaluating thermal efficiency and environmental effects. Since details on utilities consumption for the process are not given in the publications, these were roughly estimated and the necessary facilities included, together with fuel supply and environmental controls etc.

3. PROCESS DESCRIPTION

In the H-Coal process, coal is reacted catalytically with hydrogen in a slurry system to make synthetic crude. The process can also be used to make low sulfur fuel oil by operating at lower severity. For syncrude operation, reaction conditions are about 850°F and high pressure, such as 2000 psig. Syncrude production is 91,240 barrells/day for the plant feeding 25,000 tons/day of dry coal to the H-Coal reactor. An overall flowplan for the process is shown in Figure 1.

An ebullating bed reactor is used wherein the slurry of coal and catalyst in oil is agitated by bubbling hydrogen gas through it. Size of the catalyst is large relative to the coal, so that although the catalyst is fluidized, it is retained in the reactor and is not carried out with the liquid oil sidestream leaving the reactor. In addition, a gas stream is withdrawn separately from the reactor top. Further details on the H-Coal system are given in reference 15.

The following subsections describe the various operations in the overall plant. These can be conveniently grouped into several areas covering coal preparation and handling, coal liquefaction, gas separation and cleanup, liquid product recovery, hydrogen manufacture, and auxiliary facilities such as utilities, water treating, oxygen plant, and sulfur plant. This grouping will be followed through the report.

3.1 Coal Preparation and Feeding

This study assumes that cleaned coal is delivered to the plant, consequently the facilities and environmental concerns associated with coal cleaning will be at a different location, and therefore will not be covered in the present report. Coal cleaning generates considerable amounts of solid refuse to dispose of and wash water to be cleaned up for reuse as discussed in previous studies (5,11). A very large coal storage pile is included, having 30 days supply for example.

Coal feed having a nominal 10% moisture is sent first to a dryer where essentially all moisture is removed, and the coal is then crushed through 40 mesh. Crushed coal is mixed with recycle oil to form a slurry that can be pumped into the high pressure hydrogenation system. In addition, part of the dried coal goes to the gasifier so that hydrogen production can be increased to balance consumption, and dried coal also supplies the fuel used on the utility boiler.

3.2 Liquefaction Section

The coal slurry, together with makeup and recycle hydrogen, goes to a preheat furnace and then to the H-Coal reactor where hydrogenation takes place in the presence of an ebullating bed of coarse catalyst particles. About 96% of the carbon in the coal is converted to liquid or gas products, while the remaining carbon is retained in the ash which is withdrawm as a sidestream from the reactor in the form of a slurry with product oil. Part of this slurry is recirculated to the bottom of the reactor to maintain desired flow conditions.

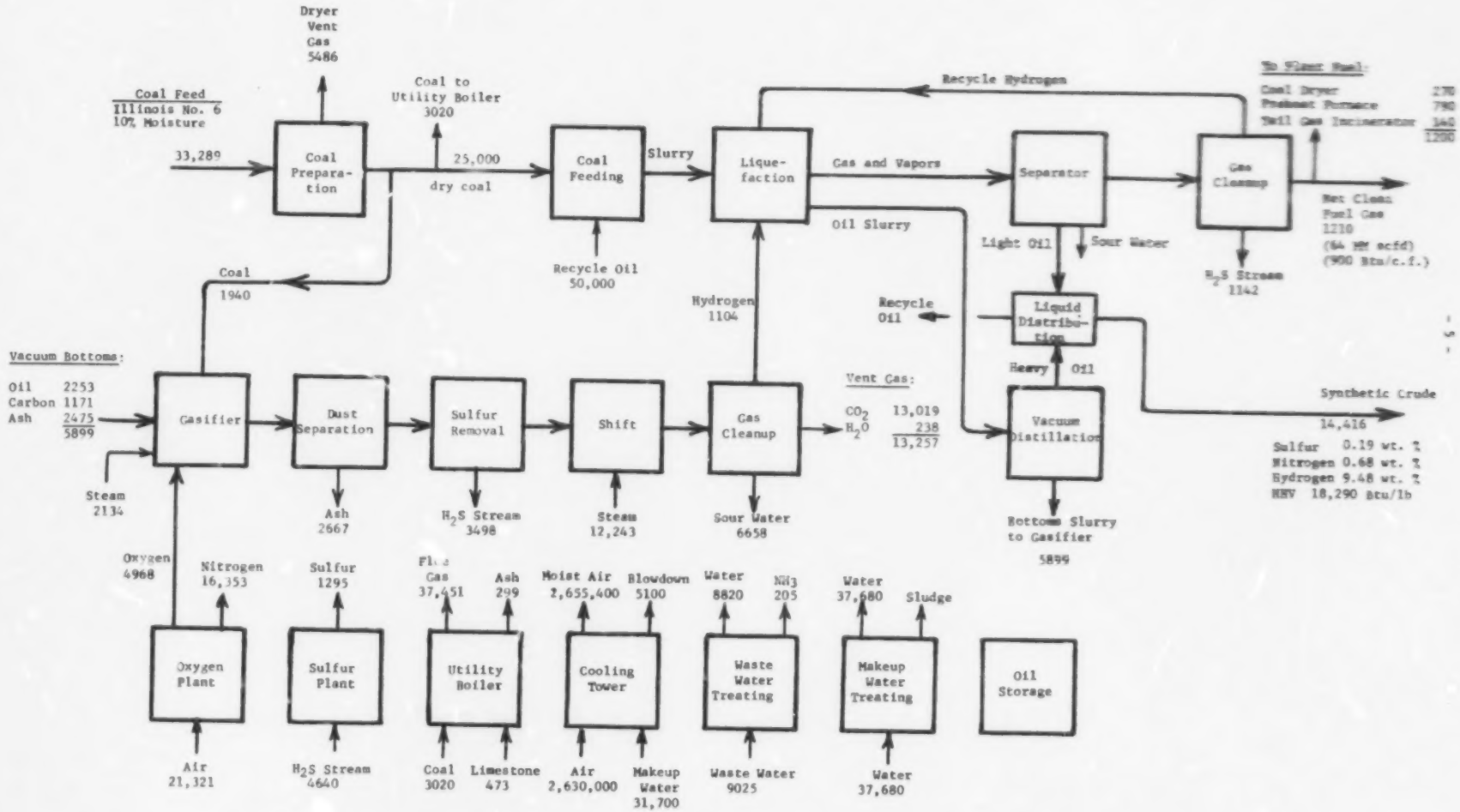


Figure 1

Block Flow Plan of H-Coal Plant for Coal Liquefaction

Note: Numbers are flow rates in tons/day.

Gases are withdrawn as a separate stream from the top of the reactor - part of the gas being recycled to the reactor inlet after cleanup to remove sulfur compounds. The remaining gas is withdrawn as a product from the process, and part of it is used to supply clean fuel to the coal dryer, reactor preheat furnace, and tail gas incinerator on the Claus plant. In the gas cleanup operation, water and oil are condensed from the gases leaving the reactor. The resulting sour water is sent to waste water treating while the oil is combined with the main liquid product.

The main oil product is withdrawn from the reactor via a liquid phase settling zone within the reactor so that the large catalyst particles are separated from the oil product and retained in the reactor. The withdrawn liquid contains ash and unreacted coal particles which are segregated by vacuum distillation into the heaviest bottom fraction of the oil. This vacuum bottoms is used to make hydrogen for the process by gasification with oxygen and steam.

Heat is recovered from the hot effluents leaving the reactor, and used to preheat feed streams or to make steam. Hydrogenation is an exothermic reaction, giving an estimated heat release for this study case of 700 MM Btu/hr, corresponding to 7700 Btu/lb hydrogen consumed, which heat is also recovered and used.

3.3 Gas Separation and Cleanup

A gas and vapor stream is withdrawn from the top of the liquefaction reactor, above the liquid level. It is substantially free of entrained liquid, and therefore contains little or no solids. Upon cooling, oil and water condense out and are separated. The sour water is sent to waste water treating, while part of the oil is recycled to form a slurry with the coal feed and the remainder of the oil is included in the final syncrude product.

The gas after condensation is cleaned up to remove sulfur compounds which are sent to sulfur recovery. Part of the clean gas is recycled to the H-Coal unit to supply hydrogen, and the rest is available as byproduct fuel gas or for plant fuel. The process used for removing sulfur from the gas is assumed to be scrubbing with an aqueous solution of amine, although hot carbonate could be used instead.

3.4 Liquid Product Recovery

A liquid stream is drawn off separately from the reactor, consisting of a slurry of ash and unreacted coal in heavy oil. This slurry is distilled under vacuum to produce a clean light distillate oil, part of which is recycled for slurrying the coal feed while the remainder is withdrawn as syncrude product along with some of the light oil condensed from the gases leaving the reactor.

Heavy bottoms from the vacuum tower, containing ash and unreacted coal, is used to make hydrogen in a partial oxidation gasifier.

3.5 Hydrogen Manufacture

A partial oxidation system is used for manufacturing hydrogen (17), consuming as raw material the slurry of vacuum bottoms which may otherwise present a disposal problem. The developer has indicated that a Texaco type partial oxidation process is used, since this type of gasifier is expected to be able to handle such a feedstock whereas some alternative processes may not be able to.

The amount of vacuum bottoms is not sufficient to make all of the hydrogen needed, so some coal feed is also sent to the gasifier, adding to the coal consumption for the plant. Oxygen for gasification is supplied by an onsite oxygen plant, while the required steam is provided from waste heat boilers. The gasification reactor operates at slagging conditions, over 2000°F, and 500 psig pressure.

Raw gas is quenched and then scrubbed with water to remove particulates including ash and soot. Water condensed at this point contains a wide spectrum of contaminants including ammonia, HCN and other nitrogen compounds, various sulfur compounds, phenols, etc., this sour water is sent to waste water cleanup.

Sulfur compounds are removed from the gas in the next processing step by scrubbing with amine. Some CO_2 is also removed but this is incidental. Amine solution from the absorber is regenerated in a stripping tower with reboiler. The sulfur containing gas stream from amine regeneration is sent to a Claus plant for sulfur recovery. Tail gas cleanup is included, as is common practice, so that the sulfur plant will meet emission requirements.

Due to the high hydrogen pressure existing in the hydroliquefaction system it is expected that sulfur in the gas will be as H_2S rather than COS, but analyses should be obtained to determine the forms of sulfur present. Hydrogen for the process is manufactured by gasification using steam and oxygen, which will no doubt result in significant COS formation. It may be desirable to provide a hydrolysis step to convert COS to H_2S and CO_2 prior to acid gas removal. This reaction is known to be catalyzed by bauxite or alumina. Alternatively hydrogen might be made by conventional steam reforming of clean byproduct gas, in which case the byproduct char and tar could be used as boiler fuel with stack gas cleanup, or the char could be gasified to make clean fuel gas.

The clean desulfurized gas is reheated and mixed with supplemental steam for processing in the shift conversion reactor. After shifting, the gas is cooled, and scrubbed to remove CO_2 using one of the available conventional systems such as hot carbonate. The CO_2 stream is vented to the atmosphere as a waste product. Environmental aspects of this stream are of particular concern, in that the flow rate is very large.

Finally, the product hydrogen is compressed and fed to the hydroliquefaction reactor which operates at about 2000 psig.

3.6 Auxiliary Facilities

The discussion so far has described the basic processing units used in a plant for hydroliquefaction of coal. In addition, auxiliary facilities are needed such as an oxygen plant, sulfur plant, and utilities systems to supply steam, electric power, and water. Waste water treating is also required. In addition to contributing effluents and emissions, these auxiliary facilities may also consume additional fuel in the form of coal or clean products from the process.

Oxygen is made by liquefaction of air, giving a waste stream of nitrogen that is clean and can be vented directly to the atmosphere. A sulfur plant is needed to recover by-product sulfur from the various sulfur compounds removed in the gas cleanup operations on the H-Coal unit and in hydrogen manufacture. A Claus type sulfur plant is used, with tail gas cleanup in order to meet environmental requirements. Total sulfur production amounts ot 1295 tons/day.

In order to make the plant self-sufficient, utility steam and electric power are generated for use in the process so that purchase of utilities is avoided. This is a basic modification of the original published case (15) in which over 200,000 KW of electric power was purchased or supplied from offsite.

Utility steam is generated at 1000 psig pressure and used to drive the turbogenerator and compressors. In some cases, bleeder turbines are used in order to balance out the generation and consumption of steam at 600 psig and 70 psig (see Table 8). Coal is used as fuel in the utility boiler, on the basis that stack gas cleanup will be provided to control emissions of sulfur and particulates. As shown in Figure 1 the amount of coal used in the boiler is 3020 tons/day on a dry basis, giving 299 tons of ash to dispose of.

Water is used for cooling, primarily to condense steam from turbines or for overhead condensers. Cooling water is recirculated at 200,000 gpm through a cooling tower where about three-quarters of the heat is dissipated by evaporation, and the remainder is taken up as sensible heat of the air passing through. The cooling tower is an area of major environmental concern in that a very large volume of air flows through the tower, and every effort should be made to see that it does not become contaminated due to leaks in exchangers, etc.

Waste water from the hydroliquefaction section, commonly called sour water, contains a wide range of pollutants including H_2S and other sulfur compounds, nitrogen compounds such as ammonia, HCN, pyridines, etc., phenols and other oxygenated compounds, plus suspended solids, oil, and tar. It would not be acceptable to discharge such water directly from the plant; therefore it is cleaned up and reused. Cleanup of waste water involves the following operations:

- Settling and filtration to remove solids.
- Extraction of phenols using a suitable solvent.

- Sour water stripping to remove H_2S, NH_3, and other low boiling materials.
- Oil removal by API type separator and froth flotation.
- Biological oxidation (biox) to consume residual small amounts of various contaminants, which are converted to cellular sludge.
- Activated carbon adsorption, if needed, for final polishing.
- Possibly special treatment for trace elements.

Ammonia will be recovered as a by-product, amounting to 205 tons/day while other contaminants removed from the waste water, such as H_2S and phenols can be sent to the sulfur plant for incineration, or returned to the process where they can be converted and destroyed.

Treated waste water is used as cooling tower makeup, supplemented by boiler blowdown and fresh water. Blowdown from the cooling tower constitutes the net water discharge from the plant amounting to 5100 tons/day (850 gpm). This blowdown, together with drift loss from the cooling tower, serves to purge dissolved solids from the system so as to prevent excessive buildup in the cooling water circuit.

Fresh water makeup is supplied to the cooling tower, as well as to boiler feed water preparation. Combined, these amount to 37,680 tons/day or 6300 gpm, which is the overall water consumption of the plant. Treating of makeup water includes lime softening and clarification, plus demineralization on the portion going to boiler feed water.

Table 1

Inputs to H-Coal Plant

Illinois No. 6 Coal Feed	Tons/Day (dry basis)
Coal to H-Coal reactor	25,000
Coal to gasifier to make H_2	1,940
Coal to utility boiler	3,020
	29,960
Makeup water	37,680

Coal Analysis, dry basis	Wt..%
Volatile matter	42.0
Fixed carbon	48.1
Ash	9.9
	100.0
Carbon	70.7
Hydrogen	5.4
Nitrogen	1.0
Sulfur	5.0
Oxygen	8.0
Ash	9.9
	100.0
High heating value, dry, Btu/lb	12,983

Table 2

Outputs from H-Coal Plant

(Based on reference 15, Table 2)

Synthetic Crude (91,240 B/D)	14,416 tons/day
Net Byproduct Gas	1,210 tons/day
Sulfur	1,295 tons/day
Ammonia	205 tons/day
Ash:	
from gasifier	2,667 tons/day
from utility boiler	299 tons/day
	2,966 tons/day
Treated waste water from plant	5,100 tons/day

Synthetic Crude Inspections

Gravity, °API	25.2
Hydrogen, wt. %	9.48
Sulfur, wt. %	0.19
Nitrogen, wt. %	0.68

Byproduct Gas

Free hydrogen, est. vol. %	56
C_1-C_3 hydrocarbons, est. vol. %	44

High heating value, ave. Btu/lb 24,000

(900 Btu/c.f.)

Yield Basis, wt. % on m.a.f. Coal

C_1-C_3 Hydrocarbons	10.7
C_4 - 400°F	17.2
400 - 650°F	26.2
650 - 975°F	18.6
975°F + Residual oil	10.0
Unreacted ash free coal	5.2
H_2O, NH_3, H_2S, CO, CO_2	15.0
	104.9

Hydrogen consumption 4.9 (18,600 scf/ton m.a.f. coal)

4. EMISSIONS TO ATMOSPHERE

All streams entering or leaving individual units of the plant are shown in the block flow diagram Figure 2 and described in Table 3. Some of these streams are returned to other processing units and thus do not leave the plant directly. Those streams that are specifically discharged to the environment are indicated by dashed lines in Figure 2 and by asterisks in Table 3. Environmental aspects of a conversion process are primarily related to emissions to the air from the plant, or effluents of liquids and solids. For discussion purposes this grouping is used and emissions to the atmosphere are covered in this section of the report.

4.1 Coal Preparation and Feeding

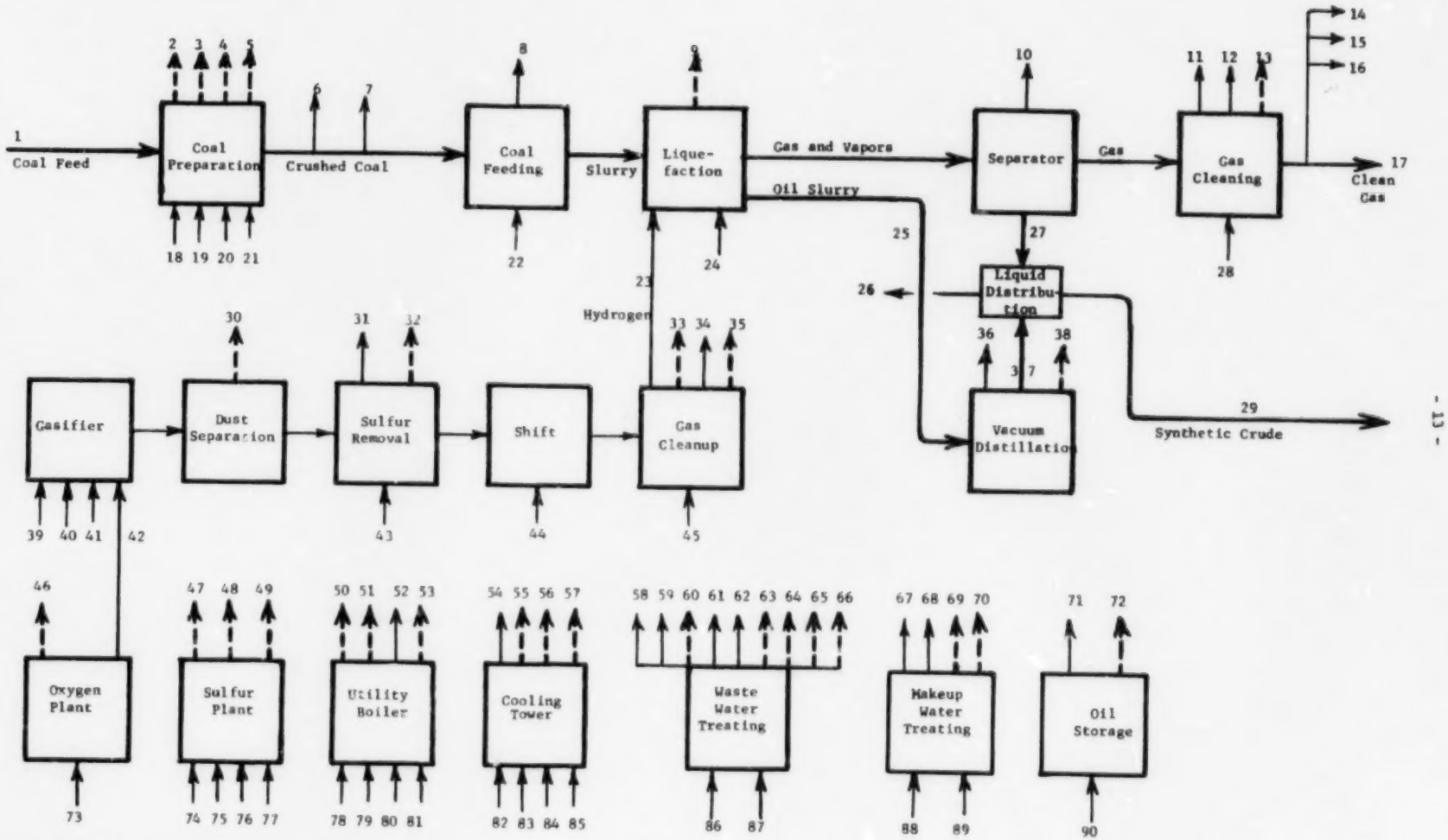
On the block flow diagram in Figure 2 the first emission to the atmsophere is from the coal storage, handling, and preparation area. Cleaned coal is delivered by rail or truck and moved to and from storage using conveyors, stackers, and unloaders. Mechanical conveyors can be a source of undesirable noise and dust, so suitable precautions should be taken. Solids handling operations will normally have a dust problem, and careful consideration and planning is required for control. Covered conveyors should be provided wherever possible; even so, there may be vent streams or leaks that could release dust. If needed, a dust collection system could be used operating at slightly below atmospheric pressure to collect vent gas and pass it through bag filters.

The coal storage pile is also of concern in that wind can pick up and disperse fine particles. Evaluation is needed for each specific situation in order to provide proper control measures. Proposals for dust control have been made such as spraying oil or asphalt on the surface of the pile, or covering it with plastic. The amount of coal handled is so large that a loss of even a small fraction of a percent could be excessive.

A further consideration on any coal storage pile is the possibility of fires and spontaneous combustion which would result in evolution of odors, fumes, and volatiles. One control measure is to compact the pile by layers as it is being formed. In any event, plans and facilities should be available for extinguishing fires if they occur (18).

It can be expected that there will be spills in the coal preparation area and that these will create a dust nuisance when they are disturbed by the wind or by trucks. Again this calls for plans and facilities for cleaning up dust and for flushing to the storm sewers.

Noise control should be carefully considered since it is often a serious problem in solids handling and size reduction. If the grinding equipment is within a building, the process area may be shielded from undue noise, but additional precautions are needed for personnel inside the building.



Note: Actual effluents and emissions to environment are indicated by heavy dashed lines; other streams are returned to process. See Table 3 for details.

Figure 2

Effluents and Emissions from H-Coal Liquefaction Plant

(Numbered streams are described in Table 3)

Table 3

Description of Streams for
H-Coal Liquefaction Plant

(see Figure 2 for numbered streams)

Stream Number	Identification	Flow Rate Tons/Day	Comments
1	Coal feed	33,289	Illinois No. 6 cleaned coal, 10% moisture. See Table 1 for details.
*2	Wind	---	Wind action in coal preparation area may cause dust problem.
*3	Rain	eg 6" in 24 hrs.	Rain runoff from coal storage area can carry suspended solids and dissolved materials.
*4	Flue gas	5486	Vent gas from dryer using clean gas fuel and 10% excess air. Dust control needed - possible odor.
*5	Coal dust	Target 0.65 max.	Residual dust in dryer vent gas leaving bag filters.
6	Coal to gasifier	1940 (dry)	Supplemental coal needed as raw material in gasifier to make all of the hydrogen used in the plant.
7	Coal fuel	3020 (dry)	Dry coal used as fuel in utility boiler.
8	Flash gas	---	Oil vapor and moisture flashed off when recycle oil is depressured and slurried with coal feed. Should be collected and returned to system.
*9	Spent catalyst	---	Rejected catalyst from liquefaction containing contaminant deposits including titania, boron, sulfur, etc. and possibly major amounts of molybdenum and cobalt.
10	Sour water	2162	Water condensate from cooling gas and vapors leaving H-Coal reactor. Will contain wide range of sulfur, nitrogen, and oxygenated compounds.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
11	H_2S Stream	1142	Concentrated H_2S stream from acid gas removal in gas cleanup sent to sulfur plant. May also contain some CO_2 and light hydrocarbons.
12	Recycle gas	---	Product gas after removing liquids and sulfur compounds, which is recycled to liquefaction reactor to supply hydrogen. Estimated H_2 content is 56 vol. %. Volume of gas recycled may be 4-5 times the makeup hydrogen rate of 420 MM scfd.
*13	Chemical	see Table 11	Purge stream of chemicals used in gas cleanup system.
14	Fuel gas	140 (542 MM Btu/hr)	Part of product gas used as fuel in coal dryer. Estimated composition is 56 vol. % hydrogen and 44 vol. % C_1-C_3. High heating value 900 Btu/c.f. or 24,000 Btu/lb.
15	Fuel gas	790	Product gas burned in reactor preheat furnace - see item 14 for details.
16	Fuel gas	270	Product gas burned in tail gas incinerator on Claus plant - see item 14 for details.
17	Product gas	1210	Net clean gas available for sale after plant fuel uses. See item 14 for gas characteristics.
18	Wind	---	Wind action on coal storage and handling area.
19	Rain	eg 6" in 24 hrs.	Rain onto coal storage and handling area.
20	Fuel gas	140	Fuel gas to coal dryer - see item 14.
21	Air	5216	Air for combustion of fuel gas to coal dryer, including 10% excess.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
22	Recycle oil	50,000	Product oil recycled to form slurry with coal feed for pumping to high pressure - based on 2 lbs oil/lb coal.
23	Hydrogen	1104	Pure hydrogen makeup to liquifaction system.
24	Recycle gas	---	Clean product gas recycled to reactor to give desired total gas flow rate - see Item 12.
25	Heavy oil slurry	---	Heavy liquid product wthdrawn as a side stream from liquifaction reactor. Contains ash and unconverted coal. Is sent to vacuum distillation to recover clean heavy distillate.
26	Recycle oil	50,000	Mixture of light oil and heavy vacuum distillate recycled for slurrying coal feed.
27	Light oil	---	Oil condensed from gases leaving liquifaction reactor - part is recycled (see item 26) and remainder is included in synthetic crude product.
28	Chemicals	see Table 11	Chemical makeup used in gas cleanup and sulfur removal (e.g. amine, etc.). May also include additives and corrosion inhibitors.
29	Synthetic Crude Product	14,416	Final clean product oil for sale or further processing. See Table 2 for inspections.
*30	Ash	2667	Ash and slag removed from raw gas leaving gasifier. May contain soot. Can be wetted for dust control.
31	H_2S Stream	3498	From sulfur removal following gasifier. Stream contains 248 tons/day H_2S in CO_2 and goes to sulfur plant.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
*32	Chemicals	see Table 11	Purge of chemicals as required to maintain capacity and activity in sulfur removal system. May contain amine, etc.
*33	CO_2 vent gas	13,257	CO_2 removed from hydrogen after water gas shift. Is discharged to the atmosphere. Includes 238 tons/day moisture.
34	Water	6658	Water condensed in gas cleanup after shift conversion and sent to waste water system.
*35	Chemicals	see Table 11	Chemical purge from scrubbing to remove CO_2 - may contain alkali carbonate and may go to waste water treating.
36	Vacuum bottoms	5899	Heavy bottoms oil and particulates remaining after vacuum distillation goes to gasification for hydrogen manufacture. See Figure 1 for composition.
37	Vacuum distillate	---	Clean light oil from vacuum tower. Part is recycled for slurrying coal feed and remainder is included in syncrude product.
*38	Vent gas	---	Small amount of flash gas removed by pump used to maintain vacuum in tower. Can go to sulfur plant or be incinerated in furnace.
39	Bottoms to gasifier	5899	See item 36
40	Coal to gasifier	1940 (dry)	See item 6
41	Steam to gasifier	2134	For gasification - based on 50% steam conversion.
42	Oxygen to gasifier	4968	Supplies all heat requirement on gasifier.
43	Chemicals	see Table 11	Makeup chemicals to sulfur removal system, which must necessarily then show up in effluents from plant.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
44	Steam	12,243	Added to give CO conversion in shift reactor.
45	Chemicals	see Table 11	Makeup chemicals for CO_2 removal e.g., alkali carbonate. May also include additives or activators.
*46	Nitrogen	16,353	Waste nitrogen from oxygen plant - vented to atmosphere.
*47	Sulfur	1295	By-product sulfur made from all collected sulfur streams, including recycle from tail gas cleanup.
*48	Tail gas	9533	Final treated vent gas from sulfur plant after incineration and tail gas cleanup.
*49	Chemicals	see Table 11	Chemical purge from tail gas cleanup process. There will also be spent Claus catalyst when it is replaced.
*50	Flue gas	37,451	Stack gas from utility boiler after stack gas cleanup.
*51	Ash	299	From coal used as fuel for steam production. May be wetted and used as landfill.
52	Steam	26,136	Steam at 1000 psig made for use in the plant so that it is self-sufficient in utilities and electric power.
*53	Spent limestone	e.g. 473	Used to remove sulfur from utility furnace flue gas. Exact form and amount will depend on operation and excess limestone used. May be in form of a water slurry.
54	Cooling water	200,000 gpm	Water from cooling tower recirculation for use in plant.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
*55	Air	2,655,400	Moist air from cooling tower includes 25,400 tons/day of evaporation from cooling water passing through tower.
*56	Mist	1200	Drift loss or spray carried out with air. Will contain dissolved solids which may cause deposits in nearby areas.
*57	Blowdown	5100	Water purged from cooling water circuit to control dissolved solids. May contain additives such as chromates, chlorine, etc.
58	Treated water	8820	Treated waste water reused in plant as makeup to cooling water. Must be sufficiently clean so that residual contaminants do not give excessive pollution of air flowing through cooling tower.
59	H_2S	---	H_2S in sour water to waste water treating is removed in sour water stripper and sent to Claus plant.
*60	Ammonia	205	NH_3 in sour water is recovered by stripping and purified for sale as a byproduct.
61	Phenols	---	Small amount of phenols may be recovered by solvent extraction and/or destroyed in biological oxidation system.
62	Oil	---	Traces of oil in sour water are removed in API type oil separator and returned to process or incinerated.
*63	Sludge	e.g. 100	Cellular material generated when contaminants are converted in biological oxidation. Some is recycled in biox unit and the net sludge product may be incinerated or possibly used as landfill.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
*64	Trace Elements	see Table 6	Considerable amounts of trace elements may be volatile and separation and/or deactivation operations may be needed - see discussion in Section 8.
*65	Chemicals	see Table 11	Various chemicals may be used in treating waste water and consequently show up in plant effluents.
*66	Evaporation	---	Ponds and settlers can lead to evaporation and odor problems.
67	Makeup water	20,720	Net fresh makeup water needed for cooling water system.
68	Boiler feed water	16,960	Makeup to boiler feed water supply after crediting recoverable condensate.
*69	Sludge	e.g. 50	From treating to cleanup water makeup to plant - e.g. lime precipitation.
*70	Backwash	---	Acid and caustic used to regenerate ion exchange resins in deminerlization to prepare boiler feed water can be combined, neutralized, and sent to waste water treating.
71	Oil	14,416	Syncrude product to sale or use. May have intended losses associat with oil storage and handling due to leaks or spills.
*72	Vapors	---	Incidental vapor release or flash ing associated with oil storage and handling could cause odor problems.
73	Air	21,321	Air used in oxygen plant to make pure oxygen for gasifier.
74	H_2S stream	4640	Gas stream containing 1390 tons/ day H_2S in CO_2, recovered from gas cleanup systems on liquefacti and on gasifier.
75	Fuel gas	140	Part of clean product gas is us in incinerator for tail gas cl up on Claus plant.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
76	Air	6048	Total combustion air for sulfur plant including tail gas incineration. See items 74 & 75.
77	Chemicals	see Table 11	Chemicals used in tail gas cleanup system.
78	Coal fuel	3020 (dry)	Coal fired to furnace generating steam needed in plant. Part of the high sulfur coal feed is used, after drying.
79	Air	34,700	Air for combustion of coal fired to utility boiler, see item 77. Includes 10% excess air.
80	Boiler feed water	26,136	Used to make steam in utility boiler. See item 51, Table 8, and Table 10.
81	Limestone	473	Used for stack gas cleanup, for example in a throwaway process. This is theoretical amount and actual use may be higher.
82	Cooling water	200,000 gpm	Cooling water returned from process to be cooled for reuse.
83	Makeup water	31,700	Total makeup water to balance blowdown, evaporation, and drift loss in cooling tower. Supplied from waste water treating (8820 tons/day) boiler blowdown (2160), and fresh water makeup (20,720).
84	Air	2,630,000	Air flow into cooling tower.
85	Chemicals	see Table 11	Additives and chemicals used in cooling water system, for example to control corrosion, fouling, or foaming.
86	Waste water	8820	Sour water etc. sent to waste water treating. See items 10 and 34.

Table 3 (Cont'd)

Stream Number	Identification	Flow Rate Tons/Day	Comments
87	Chemicals	see Table 11	Chemicals, additives, etc. used in treating waste water require consideration of possible associated disposal problems.
88	Water	37,680	Plant makeup water to be treated for use in cooling water system, and as boiler feed water.
89	Chemicals	see Table 11	Used for water treating. See comments on item 87.
90	Oil	14,416	Synthetic crude product to storage and handling. Vapors may flash off on depressuring and should be collected for return to process.

* Streams indicated by asterisk are actually emitted directly to the environment. Other streams are returned to the process.

Coal is next fed to a dryer where essentially all moisture is removed by contacting the coal with hot flue gas. To avoid releasing volatiles, temperature of coal particles at any point in the operation should not exceed 500°F. Therefore the hot combustion gases are tempered to about 1000°F by recycling cooler stack gas before being mixed with coal. Fuel efficiency is maximized by designing the heater for minimum excess air (e.g., 10%). While this increases the moisture content of the dryer offgas to perhaps 50% and tends to make the drying operation slightly more difficult, it is justified by the saving in fuel and the decreased volume of offgas to be cleaned up. In order to compensate for high moisture in the drying gas, the coal could be heated to a somewhat higher temperature, for example 210°F instead of 200°F, so as to dry to the same moisture content.

In view of high fuel costs, design of drying facilities should be reoptimized, as discussed more fully in a previous study (4). In general, it will be desirable to maximize the preheat temperature on the coal feed, and to preserve this sensible heat so as to reduce heat load on the reactor. Preheat temperatures as high as 500°F have been used without substantial evolution of volatile matter from coal but limitations such as handling and slurry pumping must also be considered.

Dust control is needed on the dryer vent gas. One conventional approach uses bag filters, with proper precautions to avoid condensation of moisture. Alternatives to consider are wet scrubbing and electrostatic precipitation. Recovered fines can be disposed of by including them in the coal slurry fed to the hydroliquefaction reactor, or the fines could be included in the coal fuel sent to the utility boiler.

Coal feed rate to the dryer is so large that a loss of only a fraction of one percent could be excessive. Thus a loss of 0.01% on coal feed would correspond to about 3 tons/day of dust in the dryer vent gas, which would result in a very noticeable dark plume. Therefore a very efficient and reliable dust control system is needed for this service.

Clean gas fuel is fired to provide heat for coal drying in this study case, since clean gas is available as a by-product. Cleanup of the dryer vent gas is thereby simplified in that sulfur removal is not needed, and bag filters should be suitable for dust control. Other fuels could be used such as coal, product oil, or some of the vacuum bottoms, although sulfur removal from the vent gas may then be needed, at least when using high sulfur coal as fuel. Fuel fired to the dryer is 542 MM Btu/hr based on removing a nominal 10 wt. % of moisture.

The dried coal is crushed through 40 mesh and sent to slurry preparation for feeding to the liquefaction system. Some dried coal also goes to gasification and to the utility boiler.

In slurry preparation the coal is mixed with recycle oil so that it can be handled and pumped to high pressure. Since the coal may contain some residual moisture, at least at times, it can be expected that steam or vapors may flash off when the coal is heated by mixing with recycle oil. Provision should be included to contain and collect any such vapors, so that they can be recovered or returned to the system and not allowed to become an effluent from the plant.

4.2 Liquefaction Section

Slurry fed to the liquefaction section at high pressure is heated to the proper temperature in a preheat furnace. Part of the clean product gas is used as fuel on the preheat furnace (1580 MM Btu/hr) so that stack gas cleanup to remove sulfur or particulates is not needed.

However, emissions of NO_x must also be defined and controlled in any specific application of the process. The amount will depend on the furnace design, use of staged combustion, fuel nitrogen content, etc. In general, NO_x production can be decreased by designing for a lower flame temperature and by using low excess air (19). Processes are being developed to remove NO_x from flue gas, and a satisfactory process will probably be available soon. These comments are also applicable to stack emissions from the coal dryer and the utility furnace.

Since the preheat furnace operates on a high pressure slurry, there is a possibility of tube failure which could result in serious emissions to the atmosphere, so consideration should be given to suitable monitoring and control techniques.

On the hydroliquefaction reactor there are no specific effluents or emissions that are intentionally released to the environment. However, this high pressure system is again subject to leaks or failures that call for very careful and thorough planning. Leaks of gases and vapors could cause objectionable odors from sulfur, phenols, etc. Spills of liquid could also cause a nuisance and might be handled by having a separate "oily water" sewer system with appropriate cleanup facilities.

4.3 Gas Separation and Cleanup

Gases and vapors withdrawn from the top of the reactor go to a cooling and recovery system. They are first cooled in exchangers to recover a maximum of useful heat. Oil vapors are thereby condensed and recovered before condensing water which might cause emulsion problems. Recovered oil is separated and withdrawn as product, or recycled for making a slurry with the coal feed to liquefaction. The gas cleanup system is enclosed, with no normal emissions to the environment except that liquids condensed at high pressure will be saturated with light gases which can flash off when the liquid is depressured. Therefore provision is needed to separate such flash gases and return them to the process, or to a furnace for incineration. Similar considerations are needed on handling all high pressure liquid streams in the plant.

Final cooling of gases from the reactor gives condensation of a large volume of sour water, which is sent to waste water treating for cleanup. This sour water will contain considerable ammonia, plus H_2S, phenols, light oil, and possibly suspended solids.

The remaining cooled gas goes to acid gas removal where sulfur compounds, particularly H_2S, are removed and sent to the sulfur plant for recovery. Some carbon oxides may be present in the gas but the amounts are small, and for this study have not been removed since fuel uses of the gas would not be affected. Unlike coal gasification, hydroliquefaction does not generate a large CO_2 stream at this point to be vented to the atmosphere. Instead, a large amount of CO_2 is vented from the system used to manufacture hydrogen. In the liquefaction route, combined oxygen in the coal feed is primarily reacted with hydrogen rather than combining with carbon, and thus appears in the sour water.

In general, the reactor section of the plant is completely enclosed and no streams are normally discharged to the atmosphere. However, the reactor operates at 2,000 psig, so that any leaks on valves or other equipment can result in serious pollution problems. For example, the air-fin coolers used on the gas and liquid products have fans to move a very large volume of air over the exchangers, and it is apparent that any leakage will be dispersed in this large air stream. Further consideration of this problem is needed to assure that the plant operations will be environmentally satisfactory (20).

Indirect heat exchange versus recirculated cooling water is also used in the high pressure reaction section as well as in other parts of the plant. It is common to find a small amount of leakage on conventional exchangers in this type of service, particularly at high pressures such as 2000 psig. Materials that leak into the cooling water can circulate to the cooling tower where they will be stripped out by the large volume of air passing through the tower. Special attention to this problem has been given in the case of oil refineries and this experience should be reviewed and applied in coal conversion operations (20,21).

Startup and shutdown of the plant, as well as maintenance, depressuring, and purging of equipment all call for special attention to control emissions. A special collection system should be used to contain and cleanup all purge and vent gas streams.

4.4 Liquid Product Recovery

As mentioned in the preceding section, some light oil is recovered from the gases leaving the reactor and is withdrawn as part of the product oil. The heavier portion of product oil leaves as a sidestream from the liquefaction reactor, and contains particulates such as ash and coal which must be removed in order to have a clean product. In this design vacuum distillation is used to make the necessary separation. Oil which is distilled overhead is clean. Part of it is recycled to use in making a slurry with the coal feed. The rest of the vacuum distillate is combined with the light oil to give the total syncrude product. Syncrude product is quite low in sulfur, 0.19 wt. %, but the nitrogen content of 0.68 wt. % is high relative to petroleum stocks. The high nitrogen content would tend to increase NO_x production if the product use involves combustion, while if it is subsequently refined the high nitrogen tends to interfere in catalytic operations.

Heat for distillation is supplied by sensible heat in the oil coming from the liquefaction reactor; consequently, no furnace is used with its attendant emissions to the atmosphere. The stream removed from the bottom of the vacuum tower contains the heaviest portions of liquid together with particulates, and is sent to hydrogen manufacture where it is gasified.

A major environmental concern on the vacuum distillation operation is the system used to maintain a vacuum in the tower. One conventional system is to use an overhead condenser operating at low enough temperature so that the vapor pressure of the distillate at the existing overhead temperature corresponds to the desired vacuum. With the heavy oil that is distilled in this design, temperature in the overhead condenser may then be roughly 100-150°F. Heat of condensation can be removed by indirect exchange to cooling water or by air cooling. Potential leaks in exchangers must be considered. In contrast to most services, the cooling water or air is now the higher pressure stream, so that any leakage will be inward into the process stream. In the case of water cooling the water leakage will mix with the oil and tend to emulsify, posing a difficult separation problem. In the case of air cooling, any air that leaks into the process stream must be removed by means of a vacuum pump which discharges at atmospheric pressure or higher. This waste gas may contain oil, sulfur, odors, etc., such that it should not be discharged directly to the environment.

A suitable way to dispose of the waste gases is by incineration, for example in the sulfur plant or in one of the plant furnaces. Even without considering exchanger leaks as a source of permanent gases that have to be rejected by the vacuum pump, there will be other sources of gases such that a typical design of any vacuum tower includes a vacuum pump downstream of the overhead condenser.

One type of vacuum pump that may be used is the ordinary mechanical design. An alternative type that is often more economical uses a steam jet ejector rather than the mechanical design. With the ejector, steam is injected at high velocity through a venturi throat which develops a low pressure zone to inspirate waste gases from the system. This is followed by an aerodynamic pressure recovery zone where the mixture of gases and steam is returned to atmospheric pressure for disposal. The steam can easily be condensed, but due to the direct contact with waste gases may be contaminated with oil, sulfur compounds, etc. This sour condensate can be included with the sour water going to waste water treating. As mentioned, the non-condensible gases can be disposed of by incineration.

4.5 Hydrogen Manufacture

Synthesis gas for use in making hydrogen is generated in a slagging gasifier where slurry bottoms from the vacuum tower plus supplemental coal are reacted with steam and oxygen. The system operates at high pressure but is entirely closed so that normally there will not be emissions to the environment. The raw gas is cooled in a waste heat boiler to make steam, followed by a water scrubbing tower to remove particulates such as

ash and soot. The water will be severely contaminated with a mixture of ammonia, phenols, sulfur compounds, etc. Up to this point in the hydrogen manufacture section, the major effluent is sour water containing residual ash and soot. In handling and disposing of these materials, emissions to the atmosphere should be avoided. The residual ash and soot might be separated from the sour water slurry by means of a settling pond or filter, to give wet ash that may be disposed of as landfill or to a mine. If allowed to dry out, ash dusting could be a problem, for example as a result of leaks or spills in the ash handling system.

Odor control may be needed on the ash and sour water streams; consequently, background information on it should also be collected during pilot unit operations. Ammonia, sulfur compounds, and phenols may all be troublesome. Facilities in a large scale plant may have to be enclosed or covered to control odors, particularly in the case of sour water. Collected vent gas can then go to suitable disposal, such as the sulfur plant or an incinerator.

In the shift converter carbon monoxide in the scrubbed gas reacts with steam to make hydrogen, plus carbon dioxide which must be removed subsequently. A fixed bed of catalyst is used and the operation is relatively clean, although the catalyst must eventually be replaced and preferably is returned to a manufacturer for reworking or refining.

After shifting, carbon dioxide is removed from the hydrogen stream using hot carbonate scrubbing for example. The CO_2 vent stream is very large, 13,257 tons/day, or roughly half as much as the total weight of coal fed to the plant. By careful design it should be possible to assure that this CO_2 stream will be essentially free of objectionable contaminants such as H_2S and COS, or combustibles. The sulfur removal system ahead of shifting should be able to give efficient sulfur cleanup, together with protection furnished by the iron based shift catalyst. Combustibles in the raw gas should be minor due to the high gasification temperature compared to some plants for making synthetic natural gas where contaminants in the CO_2 vent stream present a serious problem.

A discussion of various approaches to gas cleanup after gasification is given in reference 5, including comments on proposals for techniques to remove sulfur at high temperature so as to avoid cooling the gas more than once.

In general, scrubbing systems used to remove sulfur compounds or CO_2 will require a certain amount of chemicals makeup due to unavoidable losses, or due to side reactions that consume chemicals and require purging to maintain capacity and selectivity of the scrubbing solution (22). This results in a net effluent of chemicals from the system, and generally to the environment, which requires plans for their proper disposal. In the case of amine purge, incineration may be acceptable in that the chemical is combustible. In the case of carbonate scrubbers, the potassium carbonate cannot be destroyed by burning so some other disposal must be defined. Being water soluble, burial on land may be questionable. Ocean disposal is a possibility. Additives such as metallic complexes or inorganic salts (arsenic, vanadium, etc.) are sometimes used in scrubbing systems for acid gas removal, requiring special consideration of techniques for acceptable disposal of any chemical purges.

4.6 Auxiliary Facilities

In order to have a comprehensive and meaningful evaluation of environmental aspects of a coal conversion process, it is essential to base it on a complete plant including all related and associated facilities needed for the operation. Process fuel and utilities such as electric power should be integrated into the study, allowing for the associated increase in coal raw material consumption. The same applies to the oxygen plant, sulfur plant, and water treating facilities.

On the oxygen plant the major effluent is waste nitrogen. It should be a clean stream and can be vented to the air in a safe manner well away from structures that might be used by workers.

The sulfur plant is a potential source of considerable obnoxious emissions. An effective and reliable tail gas cleanup system is needed on the Claus plant to assure acceptably low sulfur emission. A number of processes are offered for this service and extensive commercial experience is available to draw on (23). In some of these the tail gas is first treated to reduce all sulfur compounds to H_2S which is then scrubbed out with conventional systems such as amine. In others, the sulfur compounds are first oxidized to SO_2 which is then removed by one of the techniques used for stack gas cleanup. This type of tail gas cleanup is used in the present study. Either type of process can be used to remove total sulfur in the tail gas down to a level of 250-500 ppm, corresponding to an overall sulfur separation exceeding 99% for the sulfur plant. The recovered H_2S or SO_2 is then returned to the Claus plant.

Effectiveness of a Claus plant is very sensitive to feed gas composition, especially the % H_2S in the feed (11). Processes to separate H_2S generally also separate out CO_2 at the same time, diluting the H_2S stream going to the sulfur plant. The dilution can greatly increase the volume of tail gas, requiring a correspondingly lower concentration of sulfur in the tail gas to hold a given tons/day of sulfur emissions. For example, with 25 vol. % H_2S in the gas fed to a Claus plant, tail gas volume is two times that for the same amount of H_2S at 100% concentration. At 15% H_2S the ratio is three times. The latter is more representative of a Claus plant feed stream in gasification designs using partial oxidation to make synthetic natural gas, where CO_2 in the raw gas is high relative to the H_2S content. In the present H-Coal study, feed to the Claus plant contains 35.6 vol. % H_2S with the remainder being CO_2. This favorable concentration is obtained because over 80% of the H_2S comes from gas cleanup in the liquefaction section where the H_2S is not diluted, since there is no substantial amount of CO_2 present in the gas being processed for sulfur removal.

Chemicals are used for scrubbing in the tail gas cleanup operation, leading to a chemical emission or effluent that must be recognized and evaluated for any specific case. Consumption of chemicals may either reflect physical losses such as amine vapors in the final tail gas, or the chemicals consumption may be caused by side reactions that require a purge stream in

order to maintain activity and capacity of the scrubbing solution. Thus, when scrubbing out SO_2 with sodium sulfite, some oxidation to sodium sulfate occurs. Buildup of the latter may be controlled by simply purging part of the solution, although disposal of such chemicals presents problems. Satisfactory methods need to be defined for taking care of all such chemical wastes.

The utilities section includes a boiler to provide steam and electric power. It has a large gas effluent, so that emissions of dust, sulfur, NO_x and CO must be controlled. The large fuel consumption of the boiler (3020 tons/day of coal) has a correspondingly large effect on thermal efficiency of the overall plant.

High sulfur coal is fired to the boiler equivalent to 7.6 lb SO_2/MM Btu versus the present Federal standard of 1.2 for large stationary boilers, requiring a sulfur removal of at least 84% on the stack gas. In addition fly ash will have to be removed to control dust emission, and 99% removal is needed to meet the present Federal standard of 0.1 lb particulates per MM Btu. Release of flue gases from the utility boiler is the largest gas stream that is processed and released to the atmosphere from the plant; therefore, it is particularly important to assure that emissions are controlled adequately, including transients such as at startup, etc.

Dust emission from furnaces can be controlled with demonstrated conventional equipment such as cyclones, electrostatic precipitators, or scrubbers. Sulfur can be removed as required, by one of the many processes offered for this use (24,25,26). Processes are available from the following:

Wellman-Lord	Chiyoda	FMC Corp.
Chemico	Showa Denko	Mitsui S.P. Inc.
Combustion Engineering	Babcock & Wilcox	Davy Power Gas
Universal Oil Products	Lurgi	Stauffer Chemical Co.
Research Cottrell	Enviro Chem. Systems	

Some of these are commercially demonstrated and others are undergoing large scale tests.

Emission of NO_x must also be defined and controlled in any specific application of the process. The actual NO_x formation will depend on the particular furnace design as well as the nitrogen content of the fuel fired (19,32). In general, NO_x formation can be decreased by designing for a low flame temperature and low excess air, staged combustion, and by using a fuel of low nitrogen content (19).

Although NO_x may be decreased by the above, it may still be difficult in some cases to meet the target emissions set for large stationary boilers. Considerable work is under way on methods to remove NO_x from the flue gas. While NO_2 is relatively easy to scrub out, it is found that most of the NO_x is in the form of NO which is very difficult to remove due to its low solubility in water. One answer is to convert NO to NO_2 which can then

be scrubbed out, but a simple efficient way to accomplish this is not yet available. Other approaches are to effect chemical reactions with NO_x to decompose it to free nitrogen gas. The problem is receiving intensive effort and it is expected that at least one demonstrated process will be available in the near future for use on utility boilers.

The largest volume of gas discharge to the atmosphere from the utility area is on the cooling tower. Air flow through it is about 69,000 MM cfd, and the cooling tower is therefore critical from the standpoint of pollutants. It might be expected that the recirculated cooling water would be perfectly clean and free of contaminants, however, experience shows that there will be appreciable leakage in exchangers and occasionally tube failures, especially with high pressure operations. In the present design cooling water is exchanged with oil, sour water, raw gas, amines, etc.; therefore, contaminants may get into the circulating cooling water and then be transferred to the air in the cooling tower, which necessarily provides effective contacting and stripping.

In oil refining and petrochemical operations, the cooling tower is often a major source of emissions from the plant, and techniques have been developed for making quantitative estimates of the loss (20). Control measures are also described, with emphasis on good maintenance on valves, pump seals, etc., plus floating roof tanks or vapor recovery as needed on oil and chemical storage tanks. In critical cases monitoring instruments can be provided to detect leaks.

Cooling towers also have a potential problem due to drift loss, that is mist or spray which is carried out with the air leaving the tower. Since this mist contains dissolved solids it can result in deposits when the mist settles and evaporates. Drift loss due to mist carried out with the air amounts to an estimated 1200 tons/day. New designs are being offered to reduce drift loss from cooling towers (27).

Careful consideration should also be given to the potential plume or fog problem associated with cooling towers that results from condensation under unfavorable atmospheric conditions. Condensation can occur whenever moist air leaving the cooling tower mixes with ambient air to give a mix temperature which is below that corresponding to saturation. The resulting plume can be a problem, for example, if it affects public roads. Icing of roads in the winter should also be considered. One way to prevent the plume is to provide reheat on the air leaving the cooling tower, but this results in a very large heat load and may not normally be warranted unless it can be accomplished using low level waste heat. It may be that the problems can be taken care of by proper design of the cooling tower, and by locating it in the plot plan so as to minimize impact on roads or public areas.

Waste water treating is an important area for air pollution control. Many of the contaminants in the streams to waste water treating have very strong odors so effective control measures need to be incorporated into the plant. Sour water stripping and phenol extraction are carried out in enclosed systems and should not normally have emissions. However,

it is common to use open tanks for oil separation, biological oxidation, and settling ponds which can give undesirable odors or evaporation. Careful consideration of the problem is needed and covered systems provided where appropriate.

One final consideration is the storage of oil, chemicals, sulfur, and other materials. Vapor losses can be quite significant, especially with volatile materials, due to filling and emptying tanks, or breathing due to temperature changes. Control procedures have been developed in related industries (20) and should be applied in coal conversion operations.

5. EFFLUENTS - LIQUIDS AND SOLIDS

In this part of the discussion, attention will be focused on environmental concerns related to liquid or solid streams in the plant. As in the preceding section on gas emissions, these streams are identified in Figure 2 and defined in Table 3. The order of discussion follows the processing sequence used previously.

5.1 Coal Preparation and Feeding

As mentioned earlier, the present study is based on receiving cleaned coal; consequently, the problems associated with disposing of a large amount of refuse from coal cleaning apply to some other location, together with the need for cleaning up water for reuse. Careful attention should be given to environmental aspects of coal storage and handling. Rain runoff from this area is of particular concern. Of the rain that falls onto the storage pile, some of it will run off quickly and carry suspended solids, while the remainder will soak into the pile where it will have a long contact time and can extract acids, metals, organics, etc. One approach is to collect run off water from this area in a separate sewer system and storm pond. After suitable treatment it can then provide a valuable supplement to plant makeup water.

Solids recovered in the dust collection facilities on the coal dryer can be included in feed to liquefaction or in the coal fired to the utility furnace. If coal fuel were used on the dryer instead of gas, it would contribute a residue of ash which would have to be taken into consideration in disposing of the recovered dust.

In slurry preparation the coal feed is mixed with hot recycle oil at about atmospheric pressure and perhaps 200-400°F, possibly causing some oil or moisture to flash overhead. Provision for condensing such vapors can be included, and the condensed liquid sent to the hydroliquefaction reactor or to water cleanup as appropriate.

5.2 Liquefaction Section

After pumping to high pressure, the slurry of coal feed is preheated in a furnace and fed to the hydrogenation reactor. There are no intentional effluents of liquids or solids from this section, although leaks and spills can be expected. Provision for containing these and cleaning them should be part of the planning for pollution control and housekeeping on the plant.

Maintenance and cleaning of the reactor section will require specific procedures and facilities. The reactor will contain several tons of oil slurry, which must be removed when the plant is shut down. Suspended solids will be present, as well as catalyst particles. One possibility is to store the materials removed from the reactor in suitable tanks for reuse or proper ultimate disposal. The oil is probably similar to other coal tar materials which are known to be carcinogenic, so this aspect should be evaluated and precautions taken as needed. It may be desirable to flush out the system with a neutral wash oil before opening up equipment for maintenance, although this would add to complexity.

During hydroliquefaction, some of the minor or trace elements in the coal deposit on the catalyst. Eventually, the catalyst must be regenerated or reworked. Contents of some elements in spent catalysts are summarized below (12):

Carbon	wt. % 16.4	TiO_2	wt. % 3.0
Sulfur	wt. % 4.5	Boron	wt. % 0.8
Vanadium	wt. % 0.1	Calcium	wt. % 0.5
Nickel	wt. % 0.1	Iron	wt. % 0.5

The above values were reported for Illinois coal and may be different for other coals. Although the high levels of titanium and boron in the spent catalyst may be surprising, these elements are often present in the coal feed in relatively high concentration and could be deposited on the catalyst. By way of illustration, if the catalyst replacement rate corresponds to 1 pound of catalyst per 1000 pounds of coal feed and 10 ppm of a trace element is transferred from the coal to the catalyst, then the amount deposited will give 1.0 wt. % on spent catalyst. For this illustration, catalyst makeup rate would be 25 tons/day in the plant size of Figure 1.

It is clear that specific plans are needed to handle and dispose of spent catalyst. One possibility is to return it to a manufacturer for reworking and metals recovery. If it is to be stored or buried, the extent of leaching should be defined and adequately controlled.

Obviously all materials that enter the plant, including trace elements in the coal feed must leave at some point and be disposed of in a satisfactory manner. The subject is discussed further in Section 8 on trace elements. Since much of the product oil is recycled, any trace elements collected in it will tend to build up and accumulate in this stream and may cause problems, as discussed in reference 6.

5.3 Gas Separation and Cleanup

Oil is first condensed from the gas and vapor stream leaving the reactor, at a temperature above the water dew point so as to avoid possible emulsions. This oil may contain some suspended solids or certain trace elements so pertinent information should be obtained during pilot plant operations. Environmental aspects of oil products will be discussed in Section 5.4 of this report.

After removing oil, moisture is condensed by further cooling the gas, giving a sour water stream which is sent to waste water treating. It will contain H_2S, ammonia, phenols, etc. absorbed from the gas phase.

Sour water is the major liquid stream from gas separation and cleanup. It is sent to waste water treating where contaminants are removed so that it can be reused as makeup to the cooling water circuit. The sour water will contain a wide range of contaminants including compounds of sulfur, nitrogen, or oxygen, as well as some oil and possibly solids. In addition there may be certain trace elements that could be partially vaporized in the liquefaction reactor and carried out with the gas. It can be expected that HCl and HF will tend to form from chlorides and fluorides in the coal feed when they are exposed to the high hydrogen pressure in the reactor, although there can also be reactions with ammonia, etc.

Cleanup of the sour water may require solvent extraction to remove phenols if these are present in large amounts. The phenols may then be withdrawn as a byproduct if an outlet is available, or possibly they could be recycled through hydrogenation to destroy them. Sour water stripping will remove most of the ammonia and H_2S. Ammonia may be recovered separately as a byproduct, while the H_2S should be sent to the sulfur plant.

The final step in gas cleanup is sulfur removal. This provides a clean stream of product gas that is low enough in sulfur so that it can be burned without requiring control of sulfur emission. The main effluent is a sulfur containing gas stream that is sent to a Claus plant for sulfur recovery. Concentration of H_2S separated in acid gas removal. In general, scrubbing for acid gas removal will require some makeup of chemicals which must showup as a corresponding effluent. Typically, this is a chemical purge stream, which in the case of amine scrubbing might be disposed of by incineration. An alternative to consider might be to send it to waste water treating.

The tendency of high pressure liquid streams to release gas on depressuring has been mentioned. All such flash gases should be collected and returned to some point in the process, or cleaned up before discharging.

5.4 Liquid Product Recovery

Syncrude product from the process comes from two process streams. A light portion of liquid product is recovered from the gas stream leaving the hydroliquefaction reactor, while the heavy portion is the overhead from vacuum distillation of the liquid sidestream leaving the reactor. The combined oil is syncrude product which can be further processed as desired, for example, to make motor gasoline. Based on information from other processes it appears quite likely that syncrude from the H-Coal process may contain very significant amounts of some trace elements. Thus, heavy product from the SRC coal liquefaction process has shown over 200 ppm of titanium (11), while some by-product oils from Lurgi type gasifiers are reported to contain 30-50 ppm of arsenic or lead. To the extent that such metals are contained in the H-Coal product, they may have to be recovered and disposed of in subsequent use of the product. If catalytic processing is used, the metals may deposit on the catalyst and may deactivate it. Further information on metals content of the H-Coal product is needed to clarify the situation. Solids content of the oil product is presumably very low, but should be measured.

A distinctive characteristic of oil from coal is its high nitrogen content compared to petroleum oils. Such nitrogen is known to increase NO_x formation on those fractions of the oil which find their way into fuel uses. If the oil goes to further processing, there may also be an adverse effect in that nitrogen compounds are known poisons for many catalysts. It might be thought that adsorption could be used to separate the oil molecules containing nitrogen so that they could be rejected. However, a large fraction of the oil would then be rejected. In general the nitrogen is mainly in molecules of over 300 molecular weight. If each molecule contains one nitrogen atom, its total weight will be about 20 times its nitrogen content, and there is 0.68 wt. % nitrogen in the syncrude product.

In looking at other aspects of liquid product recovery, bottoms from the vacuum tower are fed to gasification and should not contribute any new effluents in normal operation. Plant shutdown and upsets need to be considered, and it may be necessary to provide oil tanks for storing the inventory during shutdown, and for emergency use.

All storage tanks should be adequately protected to control emissions, such as vapor loss during filling, or breathing losses due to temperature changes. For oil storage, consideration can be given to floating roof tanks or completely enclosed tanks with a vapor collection system.

5.5 Hydrogen Manufacture

In hydrogen manufacture bottoms from the vacuum tower plus supplemental coal are gasified with steam and oxygen as described earlier. Raw gas is cooled and scrubbed, giving a sour water effluent which is sent to waste water treating. It contains compounds of sulfur, nitrogen, and oxygen which must be removed so that the water can be reused. Treatment of waste water will be discussed further in Section 5.6 on auxiliary facilities.

Ash remaining after gasification is collected with the sour water and has to be separated out so that it can be disposed of by burying or returning it to the mine, for example. Potential dusting and odor problems have been mentioned, but there is also a possibility of leaching contaminants from the ash by rain or ground water. Pertinent leaching information should be obtained regarding sulfur, calcium, and magnesium compounds, as well as on trace elements.

Experience shows that soot is formed in this type of gasification (17), resulting in complications in the cleanup and disposal operations. Soot production, which may be several percent on feed, is normally separated out from the water slurry and recycled to gasification but the presence of coal ash may interfere with this operation. Recycling to the gasifier is an effective way to dispose of soot, while improving thermal efficiency. Moreover, it may avoid potential disposal problems that could arise if the ash were contaminated with soot. This is an area for further study and evaluation in the various related experimental programs to define satisfactory handling and disposal methods.

It is known that many trace elements present in the coal feed are partially volatile at operating conditions used in the gasifier (28), and that considerable amounts can accumulate in the gas cleanup system - particularly in the sour water stream (29). These include many toxic elements such as arsenic, lead, cadmium, selenium, fluorine, etc. Their presence will complicate the cleanup and disposal of the sour water stream. The subject of trace elements involves special environmental problems, and is discussed in greater depth in Section 8 of this report.

After scrubbing to remove particulates, sulfur compounds are removed from the gas to prepare a clean gas for subsequent shifting. The concentrated sulfur stream goes to sulfur recovery. No major liquid or

effluents leave this unit; however, a chemical such as amine is used to scrub out sulfur compounds and normally requires a purge of chemical solution to maintain the desired capacity or activity. Provision must be made to dispose of such chemicals, possibly by incineration as mentioned in Section 5.3.

Shift conversion is the next operation, using a fixed bed of catalyst that may be of the iron type. Eventually the catalyst will need to be replaced. The spent catalyst could be returned to a manufacturer for recovery, but if it is disposed of by burying then information should be obtained to assure that secondary pollution will not be excessive due to leaching, etc.

The final cleanup of product hydrogen is by cooling to condense unconverted steam, followed by scrubbing with alkali carbonate to remove CO_2. The condensed water is relatively clean and can be used for boiler feed water makeup. As in other chemical scrubbing systems, a purge of the scrubbing solution is usually needed to prevent excessive buildup of inert or undesirable compounds resulting from side reactions or contaminants. While it might be included in the water sent to waste water treating, it would then contribute dissolved solids to the system and actually cause an increase in water consumption. A better alternative may be to return it to a chemical processer for reworking or use, possibly after evaporation to a concentrated paste.

5.6 Auxiliary Facilities

On the oxygen plant the only liquid effluent is a small amount of clean water condensed from the air, which can be used as boiler feed water makeup. On the sulfur plant the main output is byproduct sulfur which is suitable for sale. Fixed beds of catalyst are used in the Claus plant, and when replaced, can be returned to a manufacturer for disposal. Tail gas cleanup again involves scrubbing with chemicals resulting in a purge stream of chemicals that requires specific plans for disposal depending upon its exact nature.

The utility boiler burns high sulfur coal, with stack gas cleanup to control sulfur and dust emissions. Various processes are available and the form and amount of recovered sulfur compound or waste to dispose of will vary depending upon which process is selected (30,31,32,33). In one process, SO_2 in the flue gas is catalytically oxidized to SO_3 which is then absorbed in sulfuric acid and recovered as a byproduct from flue gas cleanup. Another process reacts SO_2 with oxygen in an aqueous solution of iron sulfate to form sulfuric acid, which is then neutralized with lime to form gypsum for recovery as a valuable byproduct.

For the purpose of the present study stoichiometric limestone is included for stack gas scrubbing, and could be used in a "throwaway" system, for a lime scrubber, or in a system making gypsum byproduct. The amount of limestone is 473 tpd on the above basis and introduces a considerable handling and disposal problem. Obviously it may be more desirable in many situations to have a process available that would make only pure sulfur as a byproduct.

In addition to used limestone from stack gas scrubbing there is a waste solid stream consisting of 299 tons/day of ash residue from the coal burned in the utility boiler. Consideration should be given to control of dusting, leaching, and odors in handling and disposing of all solids or waste streams.

On the cooling water system, some water must be purged in order to limit the buildup of dissolved solids. Such solids may enter in the makeup water, be introduced as chemical additives to control corrosion or algae, etc., or be formed in the process. An example of the latter would be ammonium chloride that may be formed from chlorides and nitrogen compounds present in the coal feed. A large part of the nitrogen is often converted to ammonia in hydroliquefaction or gasification, and coals often contain considerable chloride, much of which may also be released. This cooling tower blowdown represents the net water discharge from the plant.

For a typical situation the water purged from the cooling water circuit may be one-fifth of the amount evaporated in the cooling tower. Thus, the nominal concentration of dissolved solids in the purge water could then be six times that in the fresh makeup water, and may be for example over 2000 ppm such that it could be considered as brackish water and unsuitable for drinking, or even for irrigation. There are techniques for recovering clean water from such streams by evaporation, electrodialysis, etc., although there still remains the problem of how to dispose of the soluble salts or concentrated brine residue. Ocean disposal, storage, or sale are possibilities.

To inhibit corrosion chromates are often added to cooling water at concentrations of 1-10 ppm, as well as algacides such as chlorine. These interfere seriously with biological action in natural systems or in biological oxidation to remove ammonia, phenols, etc. Therefore they may have to be removed, e.g., by precipitating chromium in a pretreatment step.

There is also water blowdown from boilers but this can be used as cooling tower makeup, together with sour water which is cleaned up for reuse. Ion exchange resins are often used in demineralization to prepare boiler feedwater. Such resins are regenerated by backwashing with sulfuric acid or caustic which can then be combined, neutralized, and disposed of as discussed for cooling tower blowdown.

Waste water treating is expected to clean up the sour water so that it can be used as cooling tower makeup. The H_2S stream removed by sour water stripping together with small amounts of ammonia, hydrocarbons, etc. can be sent to the sulfur plant for incineration and disposal. In some cases byproducts such as ammonia and phenol may be recovered and sold. The flucride content of the treated waste water is of concern and should be determined in the pilot plant development. Excessively high fluoride is indicated for some large scale gasification operations, so it may be that precipitation with lime should be provided. Trace elements in addition to fluoride, such as arsenic, are expected to be present in the sour water since some are partially volatile at gasification conditions and may also be solubized in hydroliquefaction. This aspect is discussed more fully in Section 8 on trace elements.

Biological oxidation is used to clean up sour water, generating a cellular sludge which must also be disposed of. It might be suitable as a soil conditioner if satisfactory with regard to odor, trace elements, etc., or it might be disposed of by incineration. Where activated carbon is used for final water cleanup, the offgases from regeneration should be incinerated or properly disposed of. Aspects of sour water treating and cleanup are discussed more fully in reference 11.

Finally, there is treatment of makeup water which produces liquid and solid effluents. Regeneration of resins used for demineralization has already been mentioned. In addition, zeolites may be used for water softening, generating another waste stream containing dissolved solids when the zeolite is regenerated. Lime softening may be used, in which case an innocuous sludge is formed and can be disposed of along with ash residues.

6. SULFUR BALANCE

Details on sulfur balance for this study case on H-Coal liquefaction are given in Table 4. Of the total sulfur entering with the coal feed, 87.2% is recovered as byproduct sulfur from the sulfur plant. Another 8.5% is removed by stack gas cleanup on the utility boiler, resulting in a large amount of spent limestone to dispose of. Emissions to the atmosphere total 2.5% of the total sulfur, or 36 tons/day, from the boiler after stack gas cleanup plus effluent from tail gas cleanup on the sulfur plant. While the sour water contains H_2S, it will be removed by waste water treatment so that water discharged from the plant should not contain excessive amounts of objectionable sulfur compounds.

A large stream of CO_2 is vented to the atmosphere from hydrogen manufacture. It is essential that it be satisfactorily low in sulfur, which may present problems in that some of the sulfur in the raw gas from gasification is in the form of carbonyl sulfide, perhaps 10% of the total sulfur, and conventional acid gas removal systems are not very effective for removing it. One approach is to hydrolyze carbonyl sulfide and other sulfur compounds in the raw gas to H_2S, which could then be removed completely by amine scrubbing and sent to the sulfur plant. This approach has been included in the present study. After shifting, the CO_2 can then be removed using a hot carbonate type of process to give a clean CO_2 waste stream that can be vented directly to the atmosphere.

The syncrude product is low in sulfur (0.19 wt. %) and is intended for further refining and upgrading, rather than fuel uses. The process also makes byproduct fuel gas, part of which is used as plant fuel in critical services while the rest is available for sale as a clean fuel. Hydroliquefaction is expected to give only H_2S, so that complication in gas cleanup due to carbonyl sulfide should not occur in this part of the plant.

A Claus plant is used for sulfur recovery, together with tail gas cleanup to avoid excessive sulfur emissions. Sulfur compounds recovered by tail gas cleanup are returned to the Claus plant for conversion to byproduct sulfur. Overall recovery is 99% on the sulfur plant.

Table 4

Sulfur Balance H-Coal Plant

	tpd Sulfur	%
Sulfur Input		
Coal to H-Coal system	1,239	83.4
Coal to Gasifier to make H_2	96	6.5
Coal to utility boiler	150	10.1
	1,485	100.0
Sulfur Output		
Synthetic crude	27	1.8
Byproduct fuel gas	nil	--
Treated waste water	nil	--
CO_2 vent gas	nil	--
Byproduct sulfur from Claus plant	1,295	87.2
Tail gas from sulfur plant	13	0.9
Recovered from utility furnace flue gas	127	8.5
Left in utility furnace flue gas	23	1.6
	1,485	100.0

7. THERMAL EFFICIENCY

Thermal efficiency is one way to measure the effectiveness of a process for making clean products from coal. It also gives a measure of thermal pollution effects in that essentially all of the loss in thermal efficiency must be dissipated to the environment. Thermal efficiency for a process is defined as the heating value of all clean products divided by the heating value of all raw materials consumed including coal for conversion as well as coal or gas for utilities production and for plant fuel. The numbers are based on a complete and self-sufficient plant including all utilities etc., with no purchase of electricity, for example, which would make the result less meaningful.

For the H-Coal design used in this study, thermal efficiency is 75.2%. Clean products include synthetic crude and net byproduct gas as shown in Table 5, while coal is consumed in liquefaction, gasification for hydrogen manufacture, and in the utility boiler. About half of the total clean gas available is used as plant fuel on the coal dryer, reactor preheat furnace, and tail gas incinerator on the Claus plant.

As an alternative, the net byproduct gas could all be consumed within the plant by substituting it for coal used in the gasifier and/or utility boiler. Coal consumption for the overall plant would then decrease but thermal efficiency would fall to 73.2%, crediting only the synthetic crude as clean product. This number would increase somewhat if the clean gas could be used first in a combined cycle turbine for power generation, and then used in furnaces.

Some increase in efficiency might result if all hydrogen is made by conventional reforming of clean product gas, rather than from tar slurry plus coal. The oxygen plant, which is a major consumer of utilities, would then not be needed, and gas cleanup would be simplified due to the absence of sulfur and particulates. An alternative disposition of the tar slurry would be required and it might be used as fuel. One possibility is fluid bed combustion in a bed of limestone - a process that is being tested on a large pilot plant scale. Combustion with the usual stack gas cleanup might also be used, perhaps combined with a precoking step to recover oil.

Other approaches to improve thermal efficiency include possible use of heat pumps on sour water strippers or in acid gas removal. Energy savings may also be achieved by maximizing heat exchange and recovery, by more effective insulation, and by operating furnaces with lower excess air and lower stack temperatures. Suggestions for consideration are given in reference 11.

Table 5

Thermal Efficiency H-Coal Plant

	Tons/Day (Dry Basis)	10^9 Btu/Day	% of Btu
Input			
Coal to H-Coal reactor	25,000	649.2	83.4
Coal to gasifier to make H_2	1,940	50.4	6.5
Coal to utility boiler	3,020	78.4	10.1
	29,960	778.0	100.0
Output			
Synthetic crude	14,416	527.0	67.7
Net byproduct clean gas (1)	1,210	58.0	7.5
		585.0	75.2

Overall thermal efficiency: $\frac{585.0}{778.0} = 75.2\%$

(1) After providing plant fuel gas to coal dryer, reactor preheat furnace, and tail gas incinerator on sulfur plant. Sulfur byproduct has a heating value equivalent to 1.3% thermal efficiency, while byproduct ammonia is equivalent to 0.5% thermal efficiency.

8. TRACE ELEMENTS

Coal contains many trace elements present in less than 1% concentration that need to be carefully considered from the standpoint of potential impact on the environment. It is obvious that essentially all materials entering the plant must also leave via the effluent or product streams. Many of the trace elements volatilize to a small or large extent during processing, and many of the volatile components can be highly toxic. This is especially truce for mercury, selenium, arsenic, molybdenum, lead, cadmium, beryllium and fluorine. The fate of trace elements in coal conversion operations such as liquefaction or gasification can be very different than experienced in conventional coal fired furnaces. One reason is that the conversion operations take place in a reducing atmosphere, whereas in combustion the conditions are always oxidizing. This maintains the trace elements in an oxidized condition such that they may have more tendency to combine or dissolve in the major ash components such as silica and alumina. On the other hand, the reducing atmosphere present in coal conversion may form compounds such as hydrides, carbonyls or sulfides which may be more volatile. Studies on coal fired furnaces have indicated that smaller particles in fly ash contain a higher concentration of trace elements, presumably due to volatilization of these elements in the combustion zone and their subsequent condensation and collection on the fly ash particles (34,35). Other studies on coal fired furnaces are pertinent (36,37,38) and some of these report mass balances on trace elements around the furnaces (39).

Considerable information is available on the analyses of coal, including trace constituents, and these data have been assembled and evaluated (40,41,42). Very limited information is available on the fate of trace elements in the various liquefaction processes (11). A few studies have been made to determine what happens to various trace elements during gasification (2,28). As expected these show a very appreciable amount of volatilization on certain elements. As an order of magnitude, for the 29,960 tons/day of dry coal consumed by the overall H-Coal plant, each 10 ppm of trace element present in the coal contributes an input of 600 lb/day which must then leave the plant at some point. The coal is fed to liquefaction, gasification, and to the utility boiler, all of which may contribute to emission of trace elements.

Trace elements may be partially volatile in the liquefaction reactor, for example as arsine, or solubilized such that they appear in the oil or sour water. Most of the trace elements probably remain with the ash and unconverted residue and are then fed to the slagging gasifier where they are exposed to high temperature in the presence of a large volume of gas. Although only a portion of trace elements may be volatile in the gasifier, there is a very real problem to consider and evaluate since the combined amounts vaporized should be removed in the downstream gas cleaning operation and disposed of in an acceptable manner.

In order to make the picture on trace metals more meaningful, the approximate degree of volatilization during gasification shown for various elements has been combined with their corresponding concentration in a hypothetical coal (as typical), giving an estimate of the pounds per day of each element that might be carried out with hot gases. Results are shown in Table 6 in the order of decreasing volatility. Looking at the estimated amounts that may be carried overhead, it becomes immediately apparent that careful consideration of the problem is required. For each element the net amount carried overhead should be collected, removed from the system, and disposed of in an acceptable manner. In the case of zinc, boron and fluorine the degree of volatilization has not yet been determined, but they would be expected to be rather volatile. Even if only 10% of the total amount is volatile, there will be large quantities to remove in the gas cleaning operation and to dispose of.

The preceding discussion has been directed primarily at trace elements that are partially volatilized during gasification or combustion and that therefore must be recovered and disposed of in the gas cleaning systems. Consideration must also be given to trace metals that are not volatilized and leave in the solid effluents from the plant, one of which is the slag or ash from the coal fired furnace and from gasification. Undesirable elements might be leached out of this slag since it is handled as a water slurry or will ultimately be exposed to leaching by ground water when it is disposed of as land fill or to the mine. Sufficient information is not now available to evaluate the potential problems and the suituation on gasifiers may be quite different from the slag rejected from coal fired furnaces since it is produced in a reducing atmosphere rather than an oxidizing one. Background information on slag from blast furnaces used in the steel industry may be pertinent from this standpoint, since the blast furnace operates with a reducing atmosphere. However, a large amount of limestone is also added to the blast furnace, consequently the nature of the slag will be different.

Other possible sources of trace element emissions from the plant need to be evaluated. Thus, additives such as chromates may be used in the cooling water circuit and appear in the blowdown stream. Depending upon the amount present and the particular plant location, it may be desirable to provide for chromium removal, for example using lime precipitation. Similarly, trace elements may be present in chemical purge stream such as from acid gas removal systems where arsenates etc. may be used as additives, or from absorption/oxidation sulfur plants using catalysts such as vanadates. Vanadium may be an essential element in some biological systems, especially marine ones; consequently, the specific local situation will have a major effect on whether the effluent represents a problem, and on the choice of disposal method.

It is obvious that all trace elements in the coal feed must leave the plant either in the products, or in other gas, liquid, or solid effluents. It is not yet possible to make complete balances due to the early stage of process development but all data necessary for accurate and complete balances on toxic or potentially toxic elements should be obtained in the pilot plant program. Emission limitations have been specified by EPA for a number of toxic trace elements, and specifications for other elements are under consideration.

Table 6

Example of Trace Elements That May Appear in Gas Cleaning Systems

Element	Possible ppm in Coal (a)	% Volatile for Example (b)	Combined lb/day (c)
Cl	1500	>90+	80,900
Hg	0.2	90+	12
Se	2.2	74	97
As	31	65	1200
Pb	7.7	63	290
Cd	0.14	62	5
Sb	0.15	33	3
V	35	30	629
Ni	14	24	200
Be	2	18	22
Zn	44	(10)	265
B	165	(10)	989
F	85	(10)	509
Ti	340	(10)	2037
Cr	22	nil	nil

(a) Mainly based on Pittsburgh Seam Coal (2), but representative of the Illinois coal used in this study based on extensive data (42). Content of trace elements in a specific coal sample can vary considerably depending upon source, particle size, extent of cleaning, etc., as discussed in reference 42.

(b) Mainly based on a lower temperature gasifier (28) and indicated at 10% for Zn, B, and F, in absence of data.

(c) For 29,960 tons/day of coal feed total to plant including gasifier and utility boiler.

Many effluents from the plant are from conventional operations, such as process furnaces, utility boilers, waste water treating, and ash disposal. These are common, or at least similar, to other coal conversion operations or coal fired boilers, and the pollution aspects and controls have been discussed in previous reports in this series (1,2,3,4) or in other references (19,20,29,43,44,45,46).

Although elements are lost, information is needed as to where they will appear, and in what form (also vapor pressure, water solubility etc.). Such results will be needed for critical elements on all coal conversion processes used commercially, to define what recovery or separation may be required and to allow designing effective pollution control and disposal facilities. It is possible that part of the volatilized elements will enter into side reactions in the presence of sulfur, phenols, and ammonia, ash, etc., and may be soluble in water or oil, but this will not be known until further information is available.

An additional source of possible contamination from trace elements is associated with the disposal of refuse from coal cleaning. Although the cleaning operation has not been included in this specific study case, it will normally have to be provided, possibly at the mine, or in some cases at the conversion plant. It is known that sulfur compounds contained in coal refuse will oxidize upon exposure to the air and form an acid solution in the presence of water. It is quite likely that a number of trace elements can be extracted from the refuse by this acid solution. For example, similar systems have been proposed and studied for recovering copper, nickel, iron, etc. from low grade ores. It might be thought that this situation is no worse than that existing for natural mineral deposits; however, the conditions are quite different. First, the mineral has been crushed and reduced in size so that vastly more surface is exposed and available for extraction. In addition, the mineral is exposed to a large amount of oxygen, which together with the large surface area can cause considerable oxidation of sulfur compounds, organic materials, and minerals in the refuse, whereas natural mineral deposits may not be subject to such conditions. Some studies have been made in this general area (45,46) but much more work is needed.

9. TECHNOLOGY NEEDS

From this review and examination of environmental aspects of the H-Coal process, a number of areas have been defined where further information is needed in order to evaluate the situation, or where additional studies or experimental work could lead to a significant improvement from the standpoint of environmental controls, energy consumption, or thermal efficiency of the process. Items of this nature will be discussed in this section of the report, and a summary is shown in Table 7.

Any coal conversion operation has solid refuse to be disposed of. While coal cleaning is not specifically included in this study design, it will usually have to be provided at some location and necessarily generates large amounts of solid refuse to dispose of and wash water to be cleaned up for reuse. For example, there may be over 800 acre feet per year of refuse. In addition, the production of slag from gasification is 2667 tons per day or another 400 acre ft/yr. More work is needed in order to define methods of disposal that do not create problems due to leaching of acids, organic, metals, or sulfur which could contaminate natural water. In addition, adequate controls are needed with regard to the potential dust nuisance and washing away of particulates. In many cases the material may be suitable for land fill with revegetation. Although there is already a lot of background on this subject, specific information is needed on each coal and for each specific location in order to allow thorough planning to be sure that the disposal will be environmentally sound.

Coal drying is used on most coal conversion processes; consequently, considerable effort is warranted to optimize the operation from the standpoints of fuel consumption, dust recovery, and volume of vent gas to be handled (4). It will often be attractive to burn high sulfur coal rather than clean gas fuel, and to include facilities for cleaning up the vent gases.

In the liquefaction operation it will be important to determine more about what happens to various constituents in the coal feed, such as sulfur, oxygen, and nitrogen compounds and other minor or trace elements. These may be converted to materials that are soluble in the oil or water, or remain with the gas, or deposit on the catalyst. The possible formation of arsine, HF, and similar compounds should be evaluated. It is expected that much of the coal nitrogen will appear as ammonia which can be removed by scrubbing with water, but amines may also form and appear in the water layer to complicate the cleanup of waste water. Phenolic and other oxygenated compounds will probably be present. Phenol itself is very soluble in water, but higher phenols may be mainly in the oil phase. In order to clarify environmental aspects on the liquefaction operation, considerable additional information will be needed on the formation of critical minor and trace compounds. The identity and amount for each of these should be determined since it can have a major effect on the selection of cleanup and disposal methods on the oil, water, and gas streams, as well as the spent catalyst and solid wastes.

Table 7

Technology Needs

- Environmentally sound disposal of large amounts of solid refuse from coal cleaning, gasification and utility furnace, with regard to dust, leaching and sediment, trace elements, land use, etc.
- An optimized design for coal drying to use low excess air and give maximum allowable coal preheat, with good dust recovery.
- A simpler and more efficient process for acid gas removal which would provide an H_2S stream of high concentration (e.g., 50 vol. %) to the sulfur plant, while giving a separate clean stream of CO_2 that can be vented to the air. Desirable features to include:
  - good sulfur clean up, to a few ppm
  - a clean CO_2 vent stream that does not require incineration
  - low utilities consumption
  - little or no chemical purges to dispose of
- Ways to handle COS, CS_2, thiophene, etc., that are usually present in the raw gas to hydrogen manufacture and may not be removed by many acid gas removal processes. Hydrolysis to H_2S is probably one good approach.
- Sour water cleanup to make it suitable for reuse. Some purge will probably be needed to remove trace elements and perhaps ammonia and phenols. There is a great need for a practical system to evaporate sour water to make steam for use in the process, and a fluid bed system appears promising.
- Water recovery from the net water discharge leaving the plant, together with disposal of the salt concentrate.
- Information on trace elements and techniques for their disposal for liquefaction, gasification, utility furnace, and coal cleaning.
  - Extent of volatility for specific process and coal.
  - Where they appear in clean up system, and in what form. They may collect in the oil and build up by recycling. Others may appear on the hydroliquefaction or shift catalyst and in sour water or acid gas removal.
  - Many may be toxic and require separation and decontamination treatment before disposal.
  - Leaching may occur on solid wastes such as the slag or on refuse from coal cleaning. Information is needed to define the potential problem and to devise environmentally sound disposal techniques.
  - Other important discharges of trace elements must be identified for evaluation, such as chromates in cooling tower blowdown, volatile fluorides that may collect in sour water, and chemical purges from acid gas removal etc. that may contain arsenic, vanadium, etc.

In the area of acid gas removal, systems based on amine or hot carbonate are not completely satisfactory and leave room for improvement. Amine scrubbing is not effective on carbonyl sulfide, while contaminants such as cyanide interfere with regeneration of the scrubbing liquid. Hot carbonate systems do remove carbonyl sulfide, but it is often difficult to provide a highly concentrated stream of H_2S to send to the sulfur plant. In addition the CO_2 stream vented to the atmosphere may contain too much sulfur. Adsorption/oxidation systems are often not effective on carbonyl sulfide and in any event do not remove CO_2 as required, and therefore additional processing is needed. The available systems for acid gas removal have very high utility requirements, causing a significant loss in thermal efficiency for conversion of coal to clean fuel products. In addition there is often a waste stream of chemical scrubbing medium which may be difficult and expensive to dispose of. Systems based on physical solvents such as methanol appear to give a CO_2 vent stream that is excessively high in combustibles such as hydrocarbons and CO.

Desirable objectives for an acid gas removal process can be summarized as follows: (a) good clean up of all forms of sulfur to give a stream high in sulfur concentration for processing in a Claus sulfur plant, (b) effective CO_2 removal while producing a vent stream satisfactorily low in sulfur and pollutants, (c) low utility and energy consumption, (d) no waste streams that present a disposal problem.

The need for a simple, effective method to clean up sour water for reuse is another item that is common to most fossil fuel conversion operations. Sour water generally contains sulfur compounds, ammonia, H_2S, phenol, thiocyanates, cyanides, traces of oil, etc. These are generally present in too high a concentration to allow going directly to biological oxidation, but their concentration is often too low to make recovery attractive. Particulates, if present, further complicate the processing of sour water. Usual techniques for clean up include sour water stripping to remove H_2S and ammonia, and in addition, extraction may be required to remove phenols and similar compounds (11). Such operations are large consumers of utilities and have a large effect on overall thermal efficiency.

In most cases the net amount of sour water produced is less than the amount of steam consumed by reaction in gasification plus shift conversion, which suggests a way to dispose of sour water. One approach is to vaporize the sour water to make steam which can be used in the gasifier. In this case, compounds such as phenol should be destroyed and reach equilibrium concentration in the circulating sour water. It may not be practical to vaporize sour water in conventional equipment such as exchangers, due to severe fouling and corrosion problems. Therefore, new techniques may be required, and one possibility would be to vaporize the sour water by injecting it into a hot bed of fluidized solids as discussed in reference 5.

On trace elements, information is needed on the amount vaporized in the gasifier and what happens to them, where they separate out and in what form, so that techniques can be worked out for recovering or disposing of the materials. Again specific information is needed for each coal and

for each coal conversion process since operating conditions differ, particularly between liquefaction and gasification. In many cases, the trace elements may tend to recycle within the system and build up in concentration (6,29). This offers an interesting opportunity to perhaps recover some of them as useful by-products. The toxic nature of many of the volatile elements should be given careful consideration from the standpoint of emissions to the environment, as well as protection of personnel during operation and maintenance of the plant. Carcinogenicity of coal tar and other compounds present in trace amounts or formed during startup or upsets should also be evaluated. As discussed in Section 8 on Trace Elements, clarification is needed regarding potential problems associated with trace elements in various plant effluents such as spent catalyst from liquefaction and shifting or chemical purge streams from acid gas removal, tail gas cleanup, and stack gas scrubbing.

Protection of personnel, especially during maintenance operations should be given careful attention, which will require that additional information be obtained. Thus, toxic elements that vaporize in the gasifier may condense in equipment such as piping and exchangers where they could create hazards during cleaning operations.

Waste water is discharged as cooling tower blowdown, constituting the net water effluent from the plant. Detailed study of the requirements for cleaning up this water will be needed. In any event, the water make-up that is brought to the plant will contain dissolved solids including sodium and calcium salts. Calcium salts may be precipitated during the water treating operation to form a sludge which can be disposed of with the other waste solids, but the fate of the sodium salts in the make-up water calls for further study. These will leave with the blowdown from the cooling tower. If the concentration of dissolved solids is too high in this blowdown water to allow discharging it to the river, then some suitable method of disposal will have to be worked out. On one proposed commercial plant, this has been handled by using an evaporation pond where the water is evaporated to dryness. The salts accumulate and will ultimately have to be disposed of. If they cannot be used or sold then it would seem logical to dispose of them in the ocean. Other possibilities are electrodialysis, or evaporators to concentrate the salts to a paste, while recovering usable water from the waste stream.

In general it appears that there will be significant variations in emissions and effluents from different conversion operations, in addition to varying criticality of environmental factors depending upon the local situation, plant location, coal feed, etc., such that each specific plant may require its own evaluation of environmental effects and control measures.

10. PROCESS DETAILS

Additional details for the H-Coal plant including utility requirements are given in Tables 8-13.

Table 8

Steam Balance H-Coal Plant

		lb/hr
1000 psig Steam		
Generated:	Utility boiler	2,178,000
Consumed:	Electrical generation (1)	750,000
	Oxygen plant (1)	303,000
	Oxygen plant (2)	570,000
	Oxygen plant (3)	302,000
	Hydrogen compressor (3)	253,000
		2,178,000
600 psig Steam		
Generated:	Gasifier reactor	275,000
	Shift area	233,000
	H-Coal system	550,000
	Bleeder turbine	570,000
		1,628,000
Consumed:	Gasifier	178,000
	Shift reactor	1,020,000
	Acid gas removal	430,000
		1,628,000
70 psig Steam		
Generated:	Electrical generation	750,000
	Oxygen plant	303,000
		1,053,000
Consumed:	Sour water stripper	220,000
	Acid gas removal	833,000
		1,053,000

Notes: (1) Bleeder turbine discharging to 70 psig steam
(2) Bleeder turbine discharging to 600 psig steam
(3) Condensing turbine drive.

Table 9

Electric Power Required in H-Coal Plant

	kW
Coal Preparation	12,800
Scrubber	600
Acid Gas Removal	400
Gasification	400
Sulfur Plant	4,000
Slurry feed pumps	14,000
Cooling Water pumps	6,600
Cooling tower fans	3,200
Air cooler fans	4,000
Misc.	4,000
	50,000

Table 10

Water Balance for H-Coal Plant

tons/day

To Waste Water Treating - - - - - 8820

Cooling water circuit (200,000 gpm circulation):

Losses		Makeup	
Evaporation	25,400	Treated wastewater	8,820
Drift loss	1,200	From boiler blowdown	2,160
Blowdown	5,100	Fresh water	20,720
	31,700		31,700

Makeup Water

To cooling tower	20,720	
To boiler feedwater	16,960	
	37,680	(6300 gpm)

Table 11

Make Up Chemicals and Catalyst Requirements

Chemicals

Acid Gas Removal:

- scrubbing solution
- additives

Sulfur Plant tail gas cleanup

Limestone for stack gas cleanup

Cooling Tower Additives

Anticorrosion, e.g., chromate
Antifouling, e.g., chlorine

Water Treating

Lime
Alum
Caustic
Sulfuric acid

Catalysts, etc.

Liquefaction catalyst

Shift catalyst

Claus plant catalyst

Ion exchange resin for water treating

Table 12

Potential Odor Emissions

Coal storage and handling.
Coal preparation, washing, settling pond.
Coal drying - vent gas.
Vent gas from vacuum distillation.
Ash handling and disposal.
Sour water stripping and handling.
CO_2 vent stream from hydrogen manufacture.
Sulfur plant and tail gas.
Flue gas from utility boiler.
Cooling tower and air coolers.
Flash gases from depressuring liquid streams.
Biox pond and other ponds.
Leaks: ammonia, H_2S, phenols, oil, etc.

Table 13

Potential Noise Problems

Coal handling and conveyors.
Coal crushing, drying and grinding.
Oxygen plant air and oxygen compressors.
Burners and furnaces.
Stacks emitting flue gases.
Turbo-generator etc., in utilities area.
Depressuring of gases and liquids.

11. QUALIFICATIONS

As pointed out, this study does not consider cost or economics. Also, areas such as coal mining, coal cleaning, and general offsites are excluded. These will be similar and common to all conversion operations.

The study is based on a specific process design and coal type, with modifications as discussed. Plant location is an important item of the basis and is not always specified in detail. It will affect items such as the air and water conditions available, and the type of pollution control needed. For example, this study is based on high sulfur Illinois No. 6 coal, although it could be used on low sulfur western coal. Because of variations in such basis items, great caution is needed in making comparisons between coal conversion processes since they are not on a completely comparable basis.

Some other conversion processes are intended to make a clean heavy fuel, SNG, or low-Btu gas fuel, and may make appreciable amounts of by-products. Such variability further increases the difficulty of making meaningful comparisons between processes.

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